

APPEAL BRIEF - PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

J.E. Sealey II et al.

Attorney Docket No. WEYE117898

Application No.: 09/975,670

Group Art Unit: 1731

Filed:

October 10, 2001

Examiner: M.S. Alvo

Title:

PROCESS FOR MAKING A COMPOSITION FOR CONVERSION TO

LYOCELL FIBER FROM AN ALKALINE PULP HAVING LOW

AVERAGE DEGREE OF POLYMERIZATION VALUES

TRANSMITTAL OF APPEAL BRIEF

Seattle, Washington 98101 October 6, 2004

TO THE COMMISSIONER FOR PATENTS:

Enclosed herewith for filing in the above-identified application is an Appellants' Appeal Brief.

Applicants respectfully request that the shortened statutory period for filing the Appeal Brief, set to expire September 6, 2003, be extended by one month, to expire October 6, 2004.

Also enclosed is our Check No. 158540 in the amount of \$450.00, which includes the fee of \$340.00 for filing the Appeal Brief and \$110 for a one-month extension of time.

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The Commissioner is hereby authorized to charge any fees under 37 C.F.R. §§ 1.16, 1.17 and 1.18 which may be required during the entire pendency of the application, or credit any overpayment, to Deposit Account No. 03-1740. This authorization also hereby includes a request for any extensions of time of the appropriate length required upon the filing of any reply during the entire prosecution of this application. A copy of this sheet is enclosed.

Respectfully submitted,

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I hereby certify that this correspondence is being deposited with the U.S. Postal Service in a sealed envelope as first class mail with postage thereon fully prepaid and addressed to Mail Stop Appeal Brief - Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the below date.

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Applicants: J.E. Sealey II et al. Attorney Docket No. WEYE117898/23441B

Application No: 09/975,670 Group Art Unit: 1731

Filed: October 10, 2001 Examiner: M.S. Alvo

Title: PROCESS FOR MAKING A COMPOSITION FOR CONVERSION TO

LYOCELL FIBER FROM AN ALKALINE PULP HAVING LOW

AVERAGE DEGREE OF POLYMERIZATION VALUES

APPELLANTS' APPEAL BRIEF

Seattle, Washington October 6, 2004

TO THE COMMISSIONER FOR PATENTS:

This brief is in support of a Notice of Appeal filed in the above-identified application on July 6, 2004, to the Board of Patent Appeals and Interferences appealing the decisions dated March 9, 2004, of the primary Examiner finally rejecting Claims 1-11.

10/14/2004 HGUTEMA1 00000011 09975670

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I. REAL PARTY IN INTEREST

The real party-in-interest in the above-identified application is the assignee, Weyerhaeuser Company, a Washington corporation, having a place of business at 33663 Weyerhaeuser Way South, Federal Way, Washington.

II.	RELATED APPEALS AND INTERFERENCES

None.

III. <u>STATUS OF CLAIMS</u>

Claims 1-11 are pending in the application. Claims 1-11 are appealed, and all stand rejected under 35 U.S.C. § 103(a) and 35 U.S.C. § 112, second paragraph. A copy of the claims on appeal, as currently amended, is included as the Claims Appendix.

IV. STATUS OF AMENDMENTS

This application is a divisional of prior application No. 09/574,538, filed May 18, 2000, now U.S. Patent No. 6,331,354. The first Office Action was mailed on March 27, 2003. In response to the first Office Action, appellants mailed a response without claim amendments on June 10, 2003. A second nonfinal Office Action was mailed on September 16, 2003. In response to the second Office Action, appellants mailed an Amendment on December 12, 2003. The Amendment has been entered. The final Office Action was mailed on March 9, 2004. In response to the final Office Action, appellants mailed Amendment B on May 10, 2004, amending Claim 1 to its original language. Amendment B appears to have been entered, as the Advisory Action mailed on June 8, 2004, indicated the rejection of Claims 1-11 under 35 U.S.C. § 112, first paragraph, has been overcome. A Notice of Appeal was filed on July 6, 2004.

V. <u>SUMMARY OF CLAIMED SUBJECT MATTER</u>

Claim 1 is the only independent claim. All other claims depend from Claim 1. Claims 3

and 11 are argued separately. Claims 7 and 8 are argued separately. The present invention of

Claim 1 is related to a process for treating pulp to render it useful for making lyocell fibers.

Pulp is a mass of liberated fibers derived from plant material, especially trees. Depending

on the method used in producing the pulp, pulp comprises cellulose, hemicellulose, and lignin in

varying proportions. Cellulose is the predominant and most useful component of pulp. Pulp

begins as wood that is mechanically refined, chemically digested, or a combination of both

mechanical and chemical processing are used to liberate the individual pulp fibers from the

wood. Thereafter, the pulp can be chemically treated to remove undesirable coloring

components in a process referred to as "bleaching." In the most widely used chemical digestion

process, known as the "Kraft" process, wood is treated under alkaline conditions with sodium

sulfide. (Page 3, lines 22-30.) In the conventional Kraft process hemicellulose is retained within

the pulp.

In the past, cellulose was either derivatized or complexed to render the cellulose capable

of being dissolved for use in "spinning," a process for extrusion of a cellulose solution into a

regenerating bath that precipitates the cellulose as continuous filaments. (Page 2, lines 21-30.)

In recent years, attempts have been made to identify solvents that are capable of dissolving

underivatized cellulose to form dope (a solution of cellulose in water and solvent). A particularly

useful class of solvents being used today are the amine N-oxides, particularly the tertiary amine

N-oxides. With the advent of amine N-oxide solvents, an alternative now exists to the

derivatization or complexing of cellulose.

"Lyocell" is the accepted generic term for a fiber composed of cellulose precipitated from

solution without substitution of hydroxyl groups and formation of chemical intermediates.

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(Page 3, lines 1-13.) Lyocell fibers are typically produced from wood pulps that have been

extensively processed to remove non-cellulose components, especially hemicellulose. It is

generally believed that the higher the ratio of non-cellulose components present in dope, the less

suitable the dope will be for spinning into lyocell fibers. These highly processed pulps are

referred to as dissolving grade, or high alpha pulps, where the term "alpha" refers to the

percentage of cellulose. Thus, a high alpha pulp contains a high percentage of cellulose and a

correspondingly low percentage of other components, especially hemicellulose. The processing

required to generate a high alpha pulp significantly adds to the cost of lyocell fibers and products

manufactured from lyocell fibers. (Page 3, lines 14-21.)

To prepare high alpha pulps by the Kraft process, it is necessary to treat the wood with

acid before pulping. A significant amount of material, primarily hemicellulose, is solubilized in

this acid treatment. Thus, the acid treatment of pulp results in a drop in yield due primarily to the

loss of hemicellulose. (Page 4, lines 10-17.)

One measure of the suitability of a pulp useful for lyocell fiber production is its "copper"

number. The "copper" number of a pulp is proportional to the carbonyl groups present in the

cellulose. A low copper number is a desirable property of pulp that is to be used to make lyocell

fibers because it is generally believed that pulps with relatively high copper numbers degrade the

amine oxide solvent used to dissolve the cellulose. (Page 4, lines 9-16.)

Transition metals in pulp are also undesirable because transition metals tend to cause

degradation of both cellulose and solvent. (Page 4, lines 17-19.)

The present invention of Claim 1 is related to a process that can produce comparatively

inexpensive pulps as compared with high alpha pulps. The pulps made according to the process

of the invention have a high hemicellulose content, i.e., are low alpha pulps, but are nevertheless

suitable for dissolving and spinning into lyocell fibers. The pulps made by the process of the

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invention also have acceptable levels of transition metals and a relatively low copper number. Furthermore, the process of the invention can be performed using the pulps made by the conventional Kraft process without the need to perform the acid treatment step. By avoiding an acid treatment step prior to pulping, the overall cost of producing the pulp is reduced. Further, by avoiding the acid treatment, the degradation of hemicellulose is reduced and the overall yield is increased. (Page 15, lines 1-4.)

The process of the invention is performed in a reactor operated at conditions that result in a reduction of the average degree of polymerization of the cellulose in pulp. (Page 18, lines 12-18.) Degree of polymerization (D.P.) is the number of glucose monomers in a cellulose polymer. (Page 15, lines 19 to page 16, line 26.) The process furthermore does not result in any significant decrease in hemicellulose or cause a significant increase in the copper number. The invention of Claim 1 "reduces the average degree of polymerization of cellulose ..., without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number." The phrase, "without substantially increasing the copper number" means without increasing the copper number by more than about 100%, preferably not more than about 50%, and most preferably not more than about 25% during the D.P. reduction step. (Page 16, lines 15-26.) Claim 7 places a limit on the increase in copper number at less than 50%. Claim 8 places a limit on the increase in copper number at less than 50%. The phrase, "without substantially reducing the hemicellulose content" means without reducing the hemicellulose content by more than about 50%, preferably not more than about 15%, and most preferably not more than about 5% during the D.P. reduction step. (Page 15, lines 24-27.)

The present invention treats a conventional alkaline pulp, such as a Kraft pulp, having a hemicellulose of at least 7%, under alkaline conditions with an amount of an oxidant sufficient to reduce the average degree of polymerization of the cellulose to within the range of from

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about 200 to about 1100. Lowering the D.P. of the cellulose thereby results in a reduced viscosity of dope that consequently can be spun into lyocell fibers. (Page 16, lines 7-12.) The oxidant used to reduce the D.P. can be any oxidant containing a peroxide group, such as hydrogen peroxide, oxygen, chlorine dioxide, and ozone.

Prior art processes intentionally sought to remove transition metals during bleaching because it was believed that the presence of transition metals resulted in decomposition of hydrogen peroxide into cellulose-degrading intermediates that negatively impacted the viscosity of the cellulose. Unlike the prior art, appellants have discovered that they can take advantage of the presence of naturally occurring transition metals in the wood to partially degrade the hydrogen peroxide to produce intermediates that react with the cellulose to reduce its average degree of polymerization without substantially decreasing the hemicellulose content or increasing the kappa number.

Also, prior art processes have introduced magnesium sulfate as a means of inhibiting the degradation of cellulose. Unlike these processes, appellants prefer not to introduce magnesium sulfate into the reactor or upstream therefrom so that the pulp is contacted with the oxidant in the substantial absence of an inhibitor to the degradation of the cellulose. This advantage is claimed in Claim 11 that recites, "wherein the contacting step occurs in the substantial absence of an inhibitor to the degradation of the cellulose by the oxidant." If magnesium sulfate is present in the pulp prior to the reactor, it is preferred that the ratio of magnesium to the transition metals be less than 50% on a weight percent basis. (Page 20, lines 16-31.) This advantage is claimed in Claim 3 that recites, "wherein the reduction in the average degree of polymerization of the cellulose occurs in the presence of a ratio of magnesium to transition metals of less than about 50%."

The absence of an inhibitor and having reduced amounts of magnesium sulfate is an advantage in the claimed process, since the claimed process seeks to degrade the cellulose molecules by reducing the degree of polymerization of the cellulose.

VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

- (1) Whether Claims 1-11 are indefinite under 35 U.S.C. § 112.
- (2) Whether Claims 1-11 are unpatentable under 35 U.S.C. § 103(a) over U.S. Patent No. 6,042,769 (Gannon), in view of WO 99/16960 (Stephens) and U.S. Patent No. 5,985,097 (Samuelsson), with or without U.S. Patent No. 6,210,801 (Luo).

VII. <u>ARGUMENT</u>

A. Rejection of Claims 1-11 Under 35 U.S.C. § 112, Second Paragraph

Claims 1-11 are rejected under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicants regard as the invention. Appellants traverse the rejection for the following reasons.

The Examiner has stated at page 4 of the Office Action mailed on March 9, 2004 (hereinafter "Office Action") that "[t]he term 'without substantial increasing of the copper number' when read in view of the specification includes increases up to 100%. A doubling of the copper number is a substantial increase in copper number. Thus the term is indefinite. The argument that a 100% increase is not a substantial increase is not convincing. A 10% increase would be a substantial increase, a 100% increase is an extremely large increase and would not be considered 'without substantial increase.'"

The Court of Appeals for the Federal Circuit has determined that definiteness of claim language must be analyzed, not in a vacuum, but in light of (1) the content of the particular application disclosure, (2) the teachings of the prior art, and (3) the claim interpretation that would be given by one possessing the ordinary level of skill in the pertinent art at the time the invention was made. *See, e.g., In re Marosi, Stabenow, and Schwarzman*, 710 F.2d 799, 803, 218 U.S.P.Q. 289, 291 (Fed. Cir. 1983).

Appellants submit the meaning of the phrase "without substantially increasing the copper number" is not indefinite under 35 U.S.C. § 112, second paragraph. Appellants submit that the phrase "without substantially increasing the copper number" is clear to a person of ordinary skill, either based on the plain and ordinary meaning of the words themselves, and also by referring to the present specification.

The term "substantially" is often used in conjunction with another term to describe a

particular characteristic of the claimed invention. It can be a broad term. In re Nehrenberg,

280 F.2d 161, 165, 126 U.S.P.Q. 383, 386 (C.C.P.A. 1960). The limitation "to substantially

increase the efficiency of the compound as a copper extractant" was definite in view of the general

guidelines contained in the specification. In re Mattison, 509 F.2d 563, 565, 184 U.S.P.Q. 484,

486-87 (C.C.P.A. 1975). The court in Andrew Corp. v. Gabriel Electronics held that the limitation

"which produces substantially equal E and H plane illumination patterns" was definite because one

of ordinary skill in the art would know what was meant by "substantially equal." Andrew Corp. v.

Gabriel Electronics, 847 F.2d 819, 823-824, 6 U.S.P.Q.2d 2010, 2013-14 (Fed. Cir. 1988).

Words of degree, such as "substantially," are addressed in the case, Exxon v. United

States, 265 F.3d 1371, 60 U.S.P.Q.2d 1272 (Fed. Cir. 2001). In this Federal Circuit case, the

phrase "for a period sufficient to increase substantially the initial catalyst activity" did not render

the claim invalid for indefiniteness, because a person of ordinary skill would understand what the

patentee intended, even though the patent did not specify which method was used to calculate the

increase in productivity. *Id.* at 1375, 1278.

As the above cases distinctly point out, the term "substantially" is not indefinite if one of

ordinary skill in the art would understand what was meant by the phrase at issue. It is apparent

that the Examiner has understood the phrase by referring to the specification. It is, therefore,

apparent that a person of ordinary skill would also understand the phrase by referring to the

specification.

The Examiner is imposing a subjective opinion that the phrase is indefinite because the

meaning is supposedly contrary to an "accepted" meaning. See p. 4 of the Office Action mailed

on September 16, 2003, where the Examiner states "[t]he argument that Applicant is allowed to

define terms is correct. However, the definitions must be within that conventionally used in the

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The art would not recognize a doubling of copper number to be "without substantial art. increasing." However, there is no evidence in the record other than the Examiner's unsupported assertion of what is the commonly accepted meaning of "substantially." Appellants submit that there can be no prior commonly accepted meaning of the phrase because the inventive process is novel, and the phrase is one of degree. Regardless, appellants have unambiguously defined, "without substantially increasing the copper number," in the specification to be without increasing the copper number by more than about 100%, preferably not more than about 50% and most preferably not more than about 25% during the D.P. reduction step. Please see the present specification on page 16, lines 8-11. One of ordinary skill would readily understand what is meant by the phrase "without substantially increasing the copper number" by merely referring to the specification. "If the claims, read in light of the specification, reasonably apprise those skilled in the art both of the utilization and scope of the invention, and if the language is as precise as the subject matter permits, the courts can demand no more." See Hybritech Inc. v. Monoclonal Antibodies, Inc., 802 F.2d 1367, 1385, 231 U.S.P.Q. 81, 94-95 (Fed. Cir. 1986), citing Shatterproof Glass Corp. v. Libbey Owens Ford Co., 758 F.2d 613, 624, 225 U.S.P.Q. 634, 641 (Fed. Cir. 1985).

Accordingly, for all the foregoing reasons reversal of the Examiner's rejection of Claims 1-11 under 35 U.S.C. § 112, second paragraph, is respectfully requested.

B. Rejection of Claims 7 and 8 Under 35 U.S.C. § 112, Second Paragraph

Claims 7 and 8 are being argued separately. These claims are believed to be patentable for the reasons discussed above. In addition, Claims 7 and 8 place a numerical limit of the amount of increase in copper number. Appellants submit that Claims 7 and 8 are thus clear and unambiguous on the face of the claim, even without resort to the specification.

In reviewing a claim for compliance with 35 U.S.C. § 112, second paragraph, the Examiner must consider the claim as a whole to determine whether the claim apprises one of ordinary skill in the art of its scope and, therefore, serves the notice function required by 35 U.S.C. § 112, second paragraph. *See Solomon v. Kimberly-Clark Corp.*, 216 F.3d 1372, 1379, 55 U.S.P.Q.2d 1279, 1283 (Fed. Cir. 2000). Appellants submit that nothing can be more clear and precise when the metes and bounds of the claim are apparent on the face of the claim.

C. Rejection of Claims 1-11 Under 35 U.S.C. § 103(a)

Claims 1-11 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Gannon et al. (U.S. Patent No. 6,042,769) in view of Stephens (WO 99/16960) and Samuelsson (U.S. Patent No. 5,985,097) with or without Luo et al. (U.S. Patent No. 6,210,801). Appellants respectfully traverse the rejection for the following reasons.

1. Brief overview of the references.

The Gannon reference relates to a process for manufacturing a lyocell fiber having an increased tendency to fibrillate. Fibrillated fibers are advantageous for uses such as hydroentangled fabrics. Gannon describes the conventional lyocell fiber forming process that involves dissolving cellulose with amine N-oxide to form a solution, extruding the solution through a die to form a plurality of filaments, and washing the filaments to remove the solvent, thereby forming lyocell fiber. The characterizing step is described as subjecting the lyocell fiber to conditions effective to reduce the degree of polymerization of the cellulose by at least about 200 units. Col. 2, lines 2-4. Gannon describes reducing the degree of polymerization of cellulose in a lyocell fiber before or after drying, which in all cases is after the formation of the lyocell fiber. Col. 2, lines 12-22. The desired reduction in cellulose degree of polymerization may be carried out by a bleaching treatment. The bleaching treatment liquor may be applied to the fiber by passage through a bath, by padding, or by spraying. The Gannon reference does not,

however, describe how to render a pulp that is high in hemicellulose into a pulp that is useful for

spinning into lyocell fiber, without substantially increasing its copper number or decreasing its

hemicellulose content.

The Stephens reference describes a process whereby the degree of polymerization of a

pulp is reduced with a cellulolytic enzyme. The Stephens reference describes converting regular

Kraft wood pulp, without having undergone a prehydrolysis step, into a pulp suitable for the

manufacture of cellulose derivatives and lyocell fibers. The process relies on the activity of

enzymes to reduce the degree of polymerization. The process, however, removes hemicellulose.

The Samuelsson reference is related to a method that suppresses the effects of transition

and alkaline earth metal compounds during oxygen bleaching of chemically digested pulps.

According to Samuelsson, operating conditions of the oxygen bleaching stage are modified so

that depolymerization of the cellulose is low in this stage. To accomplish this objective, the

oxidation states of transition metals are changed. The change in oxidation state can be brought

about by changes in the oxygen pressure, alkali concentration and temperature.

The Luo reference is related to a process for reducing the degree of polymerization of

cellulose of a pulp using a variety of chemicals. The pulps produced thereby are useful for

making lyocell fibers. One of the advantages of lyocell fibers produced by the pulps made

according to the Luo reference is the decreased tendency of the lyocell fibers to fibrillate.

According to the Luo process, after reducing the degree of polymerization of cellulose, the

copper number of the pulp has to be reduced with sodium borohydride or sodium hydroxide.

The Examiner suggests the claimed invention is obvious by combining the Gannon,

Stephens, Samuelsson and Luo references.

An obviousness rejection requires that there be a suggestion or motivation either in the

references or in the knowledge that is generally available to modify a reference or to combine

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references. In addition, there must be a reasonable expectation of success and all the elements of the claims must be taught or suggested by the prior art.

Appellants will show that the Examiner has failed to provide all the elements of a *prima* facie case of obviousness.

2. There is no suggestion or motivation to combine the Stephens reference with the Gannon reference. There is no reasonable expectation of success. The references do not describe all the elements of the claimed invention.

From the brief overview of the Gannon reference above, it is evident that the Examiner has mischaracterized the Gannon reference by suggesting pulp and lyocell fiber mean the same thing, as is evident from the Examiner's statement on page 3 of the Office Action:

Gannon et al. fairly disclose a process for making lyocell fibers comprising the steps of: (a) contacting an alkaline pulp comprising cellulose and hemicellulose under alkaline condition with an amount of oxidant (hydrogen peroxide or ozone) sufficient to reduce the average degree of polymerization of the cellulose to the range of from about 200 to about 1100 and (b) forming the fibers from the pulp treated in accordance with step (a) (see Cols. 2-6). Gannon et al. teaches the claimed invention except for the limitation of without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number.

There is no indication by the Examiner where the Gannon reference specifically describes steps (a) and (b), other than to make broad reference to Columns 2 through 6.

Claim 1 recites "contacting an alkaline <u>pulp</u> comprising cellulose and at least about 7% hemicellulose under alkaline conditions with an amount of an oxidant sufficient to reduce the average degree of polymerization of the cellulose to within the range of from about 200 to about 1100, without substantially reducing the hemicellulose content of the <u>pulp</u> or substantially increasing the copper number."

In direct contrast to Claim 1, Gannon describes a lyocell <u>fiber</u> treatment that reduces the lyocell <u>fiber's</u> degree of polymerization by about 200 units. See the Gannon reference's Abstract.

A lyocell fiber is described in the SUMMARY OF CLAIMED SUBJECT MATTER section of this appeal brief as being precipitated (i.e., regenerated) from a solution of cellulose, water and a solvent. In direct contrast, pulp that is the object of the present inventive process is the precursor material of lyocell fibers that has not undergone dissolution and consequently is not regenerated.

It is the formed lyocell fibers that are described as being treated by the Gannon reference, not the

pulp as claimed. (Col. 2, lines 12-22.)

response, mailed on May 10, 2004.

Appellants believe the Examiner's reading of "pulp" in the claims being synonymous with the "lyocell fiber" of Gannon, is in error. Not only do the words themselves define different materials. It can be shown that there are structural differences between the cellulose in pulp and the cellulose in a lyocell fiber. Consequently, a "pulp" and a "lyocell fiber" are recognized in the art as being materially different. In support of appellants' position that pulp and lyocell fiber are structurally different, appellants direct the attention of the Board to two excerpts of publications included in the evidence appendix of this appeal brief. The excerpts were included with a

At page 53 of the Sjöström publication, the differences of Cellulose I, which is native cellulose, are contrasted with regenerated cellulose, Cellulose II. Furthermore, on page 55, the publication states, "Cellulose II is formed whenever the lattice of Cellulose I is destroyed, for example, on swelling with strong alkali or on dissolution of cellulose." Appellants submit that the Gannon reference describes the treatment of Cellulose II because the Gannon reference explicitly describes the dissolution of cellulose in a solvent followed by regeneration and treatment (Col. 1, line 65). See also the Rydholm publication, at pages 116-17, describing the differences in cellulose between native cellulose (Cellulose I) and regenerated cellulose (Cellulose II). Again, lyocell fiber is a regenerated cellulose.

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Accordingly, based on the discussion above, it is not correct to state that Gannon et al. disclose contacting an alkaline <u>pulp</u> under alkaline conditions with an amount of oxidant, when, in fact, what is described by Gannon is treatment of a <u>lyocell fiber</u>. Pulp and lyocell fiber do not mean the same thing.

Furthermore, the Gannon reference is silent on the amount of hemicellulose present in the lyocell fiber and therefore cannot teach the reduction of the D.P. of cellulose without substantially decreasing the hemicellulose content of a pulp with at least 7% hemicellulose. This alone distinguishes the claimed invention over the Gannon reference.

Perhaps appreciating that the Gannon reference does not describe treating a pulp, the Examiner further states that, "[t]he argument that Gannon uses a different starting material than the instant process is not convincing as it would have been obvious to use non-regenerated cellulose from the teachings of WO 99/16960 [Stephens]," and "[i]t would have been *prima facie* obvious from the teachings of WO 99/16960 [Stephens] to use alkaline Kraft pulp as the fiber of Gannon et al. The Kraft pulp of WO 99/16960 [Stephens] is the same non-regenerated cellulose used by Applicant."

Before the references can be combined or modified there has to be a suggestion or motivation to do so. "Obviousness cannot be established by combining the teachings of the prior art to produce the claimed invention absent some teaching, suggestion or incentive supporting the combination." *Ecolochem, Inc. v. Southern California Edison*, 227 F.3d 1361; 1372, 56 U.S.P.Q.2d 1065, 1073 (Fed. Cir. 2000), *citing ACS Hosp. Sys., Inc. v. Montefiore Hosp.*, 732 F.2d 1572, 1577, 221 U.S.P.Q. 929, 933 (Fed. Cir. 1984). If the motivation is not immediately apparent, it is the duty of the Examiner to explain why the combination of the teachings is proper. *Ex parte Skinner*, 2 U.S.P.Q.2d 1788, 1790 (Bd. Pat. App. & Inter. 1986).

Broad allegations of obviousness and of the teachings of Stephens reference do not further prosecution. No explanation is provided why the modification of Gannon with Stephens would be obvious. Furthermore, any attempt at combining and/or modifying the Gannon reference with the Stephens reference makes no sense. The Gannon reference is directed at treating lyocell fibers. The Stephens reference is directed at treating pulp.

Is the Examiner implying that prior to forming the lyocell fibers, Gannon should treat the pulp? Or is the Examiner implying that Stephens could substitute bleach liquor for enzymes? Either way, modification of the Gannon or Stephens reference involves a complete change in the principle of the operation of the Gannon and Stephens inventions. The Gannon reference's objective is to produce a lyocell fiber with an increased tendency to fibrillate. See Col. 1, lines 61-64. Appellants submit the Gannon reference teaches away from reducing the D.P. of pulp. Gannon mentions that the cellulose concentration in dope prior to spinning is directly proportional to the tendency of lyocell fibers to fibrillate. However, increasing the cellulose concentration requires reducing the D.P. of cellulose to maintain the dope viscosity below the maximum working viscosity. Gannon does not see this as a viable alternative to his method. The increase in fibrillation tendency achievable by use of the Gannon process is generally greater than the increase achievable by raising the cellulose concentration of the solution. (See Col. 5, lines 1-4).

The combination of Gannon with Stephens does not result in the claimed invention. If one were to combine Stephens with Gannon, the pulp used by Gannon would be substituted with the pulp used by Stephens. The pulp would then be dissolved in a solvent, and extruded. However, as taught by Sjöström, the dissolution process would result in cellulose II fiber before it was treated by the process of Gannon. Once again, as stated at the very beginning of this

section, lyocell fiber and pulp are not the same. Therefore, combination of Gannon with Stephens does not result in the claimed invention.

As discussed above, the Gannon reference is specifically directed at treating the formed lyocell fibers, not the pulp. Combining or modifying the Gannon reference with the Stephens reference would not only change the principle of operation of the Gannon reference, but also would appear to render the Gannon reference unsatisfactory for its intended purpose. If the Stephens method is modified by incorporating the Gannon method, that is a complete revision of the Stephens invention. If the proposed modification renders the prior art invention being modified unsatisfactory for its intended purpose, there is no suggestion or motivation to make the proposed modification. *In re Gordon*, 733 F.2d 900, 902, 221 U.S.P.Q. 1125, 1127 (Fed. Cir. 1125). If the proposed modification or combination of the prior art would change the principle of operation of the prior art invention being modified, then the teachings of the references are not sufficient to render the claims obvious. *In re Ratti*, 270 F.2d 810, 813, 123 U.S.P.Q. 349, 352 (C.C.P.A. 1959).

Finally, the invention defined by Claim 1 recites that the pulp that undergoes the D.P. reduction process has at least a 7% hemicellulose content that is not substantially reduced at the time of reducing the D.P. of the cellulose. The combination of the Gannon and the Stephens references fails to teach a process for reducing the D.P. of cellulose that also does not result in any substantial reduction in hemicellulose content of a pulp having at least 7% hemicellulose or any substantial increase in copper number. Contrary to the invention defined by Claim 1, the Stephens reference describes the removal of hemicellulose, not an attempt at avoiding its reduction. See page 4, lines 15-17; page 8, lines 9-11; and page 8, lines 19-21 in the Stephens reference.

Because at the very least, there is no suggestion or motivation to combine the Stephens reference with the Gannon reference, no reasonable expectation of success, and a clear lack of all of the elements of Claim 1, the Examiner has not established a *prima facie* case of obviousness. The deficiencies in the Gannon and Stephens references are not cured by either Samuelsson, Luo, or both as further discussed below. Accordingly, the reversal of the Examiner's rejection of Claim 1 and dependent Claims 2-11 is respectfully requested.

3. There is no suggestion or motivation to combine the Samuelsson reference with the Gannon reference. There is no reasonable expectation of success. The references do not describe all the elements of the claimed invention.

At page 3 of the Office Action, the Examiner states:

Samuelsson et al. teaches that the catalysation of the depolymerization of cellulose and hemicellulose during peroxide bleaching can be controlled by monitoring and controlling the ratio of transition metals and Mg in the pulp (Col. 4, lines 10-22 and 45-47, of the Samuelsson reference). It would have been obvious to prevent the degradation of the hemicellulose and cellulose in the cellulosic material of Gannon et al. by adding the proper amount of Mg as taught by Samuelsson et al. Obviously such addition would avoid substantial reduction of the hemicellulose content, which would not be degraded.

The Samuelsson reference is directed to a modification of an oxygen bleaching process to prevent the depolymerization of cellulose with the addition of magnesium and manganese. See Col. 2, lines 37-47. The only reference to hemicellulose appears in Col. 1, line 35. This brief mention of hemicellulose only addresses the issue that metals in pulp leads to a depolymerization of hemicellulose. The Samuelsson reference never teaches whether degradation of hemicellulose could or should be prevented. The Samuelsson reference's main focus is directed towards preventing the depolymerization of cellulose. See Col. 3, lines 24-27.

The Samuelsson reference's objective of preventing the depolymerization of cellulose is directly contrary to the objective of Gannon which is to reduce the degree of polymerization of

cellulose to increase the tendency of a lyocell fiber to fibrillate. There is no reason why Gannon should prevent depolymerization of cellulose when the explicit teaching of Gannon is to cause depolymerization of the cellulose. (See Col. 2, lines 3-5.) Accordingly, there is nothing to suggest or motivate one to combine or modify Gannon in a way based on Samuelsson.

Also, in direct contrast to the Samuelsson reference, Claim 1 specifically recites reducing the degree of polymerization of cellulose . . . without substantially reducing the hemicellulose content or increasing the copper number. Not only is there no suggestion or motivation to combine the Samuelsson reference with the Gannon reference, but any attempt to do so is bound to fail because implementing the suggestion to prevent the degradation of hemicellulose by adding magnesium will necessarily also lead to preventing the degradation of cellulose. The prevention of cellulose degradation is both contrary to the Gannon reference and Claim 1. In the SUMMARY OF THE CLAIMED SUBJECT MATTER, appellants have pointed out that the prior art teaches the introduction of magnesium sulfate as a means to inhibit the degradation of cellulose, whereas appellants find it advantageous not to introduce magnesium sulfate, and further describe that if magnesium sulfate is naturally present in the pulp, its content is limited to less than a specific ratio. The object of the Gannon reference is to decrease the degree of polymerization of cellulose, not suppress cellulose depolymerization which is what will occur if magnesium compounds are added, as taught by Samuelsson. Consequently, adding magnesium to the method described by Gannon leaves this prior art invention inoperative. When the prior art teaches away from the claimed invention, the invention is not obvious in view thereof. In re Sponnoble, 405 F.2d 578, 588, 160 U.S.P.Q. 237, 244 (C.C.P.A. 1969) ("a combination of [references] would produce a seemingly inoperative device." Parenthetical added).

There is no suggestion or motivation to combine or modify Gannon based on Stephens.

There is no reasonable expectation of success by combining Gannon with Stephens. The

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combination of Gannon with Stephens fails to provide all elements of the claimed invention.

The suggestion or motivation to combine or modify Gannon with Stephens and Samuelsson is

not found in Samuelsson. Samuelsson teaches away from combining with Gannon. The

combination of Samuelsson with Gannon and Stephens does not yield a reasonable expectation

of success nor does the combination of Gannon, Stephens and Samuelsson provide all the

elements of the claimed invention.

4. There is no suggestion or motivation to combine the Luo reference with the

Gannon reference. There is no reasonable expectation of success. The references

do not describe all the elements of the claimed invention.

At pages 3-4 of the Office Action, the Examiner states that,

[s]ince the copper number is directly related to the cellulose degradation,

see specification, page 16, lines 8-20, it would have been obvious that preventing cellulose degradation by adding Mg to the pulp during peroxide bleaching as taught by Gannon et al., would prevent an increase

in the copper number.

Before appellants address the errors with combining the Gannon and Luo references,

appellants wish to correct what appears to be a misunderstanding on the part of the Examiner.

The passage taken from appellants' specification (page 16, lines 8-20) is in the context of

dissolving the cellulose in the amine oxide solvent. That passage relates to the degradation of the

solvent that can occur during and after dissolution of the pulp to form a dope. The passage does

not state that a direct relationship exists between the degree of polymerization and the resultant

copper number.

There is no teaching from the references so far discussed above to reduce the degree of

polymerization of cellulose without substantially increasing the copper number at the time of

reducing the degree of polymerization.

The Examiner states at page 4 of the Office Action that:

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[i]f necessary, it would have been obvious to prevent an increase in the copper number by treating the lyocell with sodium borohydride to decrease the copper number as taught by Luo et al.

Again, a broad allegation that something is obvious is meaningless without a suggestion or motivation to combine references. There is no suggestion or motivation to combine the Gannon reference with the Luo reference.

The Luo reference describes the reduction of the D.P. of cellulose of pulp, followed by the reduction in copper number. As explained in THE SUMMARY OF THE CLAIMED INVENTION, a low copper number is a desirable characteristic in a pulp that is to be dissolved, because a pulp with a relatively high copper number degrades the solvent. Appellants fail to see the value of reducing the copper number of Gannon's lyocell fibers when the fibers have already been dissolved, spun and formed.

Even if the Luo and Gannon references are combined, the resultant process is not the claimed process. Claim 1, paraphrased in part, requires prevention of a substantial increase in copper number during the treatment to reduce the degree of polymerization. See the SUMMARY OF THE CLAIMED INVENTION. Luo describes a two-step process. The first step reduces the D.P. of the cellulose of the pulp. The second step reduces the copper number of the cellulose of the pulp. The second step is necessary in the Luo reference because the reduction in the D.P. of cellulose causes an increase in copper number. See Col. 13, lines 31-39, of the Luo reference. Appellants' claimed invention eliminates the need to have a separate, distinct step for reducing the copper number. Accordingly, the Luo et al. reference does not describe a step to "reduce the average degree of polymerization... without... substantially increasing the copper number," as recited by Claim 1. Elimination of an element (or step) with retention of its function is an indicia of unobviousness. *In re Edge*, 359 F.2d 896, 899, 149 U.S.P.Q. 556, 557 (C.C.P.A. 1966).

Furthermore, one of the advantages described by Luo, is the decreased tendency of lyocell fibers to fibrillate. See the Abstract. The Gannon reference's objective is to increase the

fibrillation tendency of lyocell fibers. Accordingly, there is no suggestion or motivation to

combine the Luo reference with the Gannon reference.

There is no suggestion or motivation to combine or modify Gannon based on Stephens

and Samuelsson. There is no reasonable expectation of success by combining Gannon with

Stephens and Samuelsson. The combination of Gannon with Stephens and Samuelsson fails to

provide all the elements of the claimed invention. The suggestion or motivation to combine or

modify Gannon with Stephens, Samuelsson and Luo is not found in Luo. Luo teaches away

from combining with Gannon. The combination of Luo with Gannon, Stephens and Samuelsson

does not yield a reasonable expectation of success nor does the combination of Gannon,

Stephens, Samuelsson and Luo provide all the elements of the claimed invention.

D. The Rejection of Claims 3 and 11 Under 35 U.S.C. § 103(a)

Claims 3 and 11 are being argued separately. These claims are believed patentable for

the reasons discussed above. In addition, Claim 3 recites, "wherein the reduction in the average

degree of polymerization of the cellulose occurs in the presence of a ratio of magnesium to

transition metals of less than about 50%" and Claim 11 recites, "wherein the contacting step

occurs in the substantial absence of an inhibitor to the degradation of the cellulose by the

oxidant."

As explained in the SUMMARY OF THE CLAIMED INVENTION, appellants have

found it advantageous not to add magnesium sulfate or other inhibitor that prevents the

depolymerization of cellulose. The reason for not having an inhibitor or having a limit on the

amount of magnesium is because the claimed invention seeks to advantageously reduce the D.P.

of cellulose of a pulp to within a range that can produce a dope that can be spun into lyocell

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fiber. Any inhibitor, or magnesium compound would not allow the reduction of the D.P. of cellulose. There is no teaching or suggestion in the references discussed above to perform a method in accordance with either Claims 3 or 11. In direct contrast, the only reference that appears to address the issue of magnesium is the Samuelsson reference. However, the Samuelsson reference describes the addition of magnesium compounds (inhibitors) to prevent depolymerization. Appellants reduce or eliminate the amount of magnesium and inhibitor to achieve the opposite result, a reduction in the D.P. of cellulose. Accordingly, Claims 3 and 11 are patentable in view of Gannon, Stephens, Samuelsson, and Luo, taken alone or in

E. The Rejection of Claims 7 and 8 Under 35 U.S.C. § 103(a)

Claims 7 and 8 are being argued separately. These claims are believed patentable for the reasons discussed above. In addition, Claim 7 recites "wherein the copper number increases less than 50%," and Claim 8 recites, "wherein the copper number increases less than about 25%."

Of all the references that were cited and applied, the only reference that addresses copper number is the Luo reference. However, the Luo reference describes that the increase in copper number cannot be prevented during the D.P. reduction step, and finds it necessary to perform a second and subsequent step to reduce the copper number. In contrast, the invention defined by Claims 7 and 8, advantageously holds down the copper number increase to a small percentage that avoids the need for a second subsequent step to reduce the copper number. Accordingly, Claims 7 and 8 are patentable in view of Gannon, Stephens, Samuelsson, and Luo, taken alone or in combination.

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combination.

VIII. **CONCLUSION**

In view of the above remarks, appellants respectfully submit that each of Claims 1-11 is patentable over the references of record. A decision reversing the Examiner's rejections and finding all pending claims to be in condition for allowance is respectfully requested.

Respectfully submitted,

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I hereby certify that this correspondence is being deposited in triplicate with the U.S. Postal Service in a sealed envelope as first class mail with postage thereon fully prepaid and addressed to Mail Stop Appeal Brief -Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the below date.

Date:

LXC:gm/jlj

CLAIMS APPENDIX

1. (Previously presented) A process for making a composition for conversion to lyocell fiber, said process comprising:

contacting an alkaline pulp comprising cellulose and at least about 7% hemicellulose under alkaline conditions with an amount of an oxidant sufficient to reduce the average degree of polymerization of the cellulose to within the range of from about 200 to about 1100, without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number.

- 2. (Original) The process of Claim 1 wherein said oxidant comprises at least one member of the group consisting of a chemical with a peroxide group, oxygen, chlorine dioxide, ozone and combinations thereof.
- 3. (Original) The process of Claim 2 wherein the reduction in the average degree of polymerization of the cellulose occurs in the presence of a ratio of magnesium to transition metals of less than about 50%.
- 4. (Original) The process of Claim 1 wherein the hemicellulose content of the pulp is reduced less than about 50%.
- 5. (Original) The process of Claim 4 wherein the hemicellulose content of the pulp is reduced less than about 15%.
- 6. (Original) The process of Claim 4 wherein the hemicellulose content of the pulp is reduced less than about 5%.
- 7. (Original) The process of Claim 1 wherein the copper number increases less than 50%.
- 8. (Original) The process of Claim 1 wherein the copper number increases less than about 25%.

- 9. (Original) The process of Claim 2 wherein the contacting step further comprises contacting the pulp with an alkali source selected from the group consisting of sodium hydroxide, oxidized white liquor, and unoxidized white liquor.
- 10. (Original) The process of Claim 1, wherein the alkaline pulp and oxidant are contacted at a pH greater than about 8.0.
- 11. (Original) The process of Claim 1, wherein the contacting step occurs in the substantial absence of an inhibitor to the degradation of the cellulose by the oxidant.

EVIDENCE APPENDIX

- A. Rydholm, Sven A., *Pulping Processes*, InterScience Publishers, 1965, pp. 112-119.
- B. Sjöström, Eero, Wood Chemistry: Fundamentals and Applications, Academic Press, 1981, pp. 52-57.

Pulping Processes

SVEN A. RYDHOLM

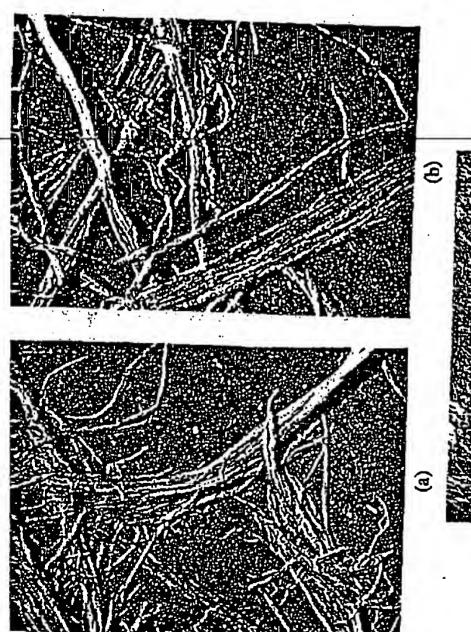
Research Director Billeruds AB, Säffle, Sweden

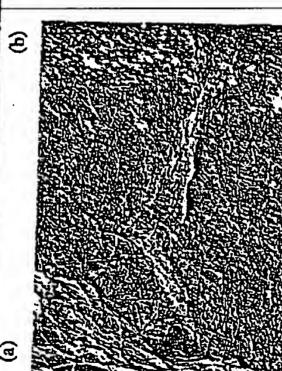


ERIK HÄGGLUND [1887–1959] Pioneer of modern pulping chemistry

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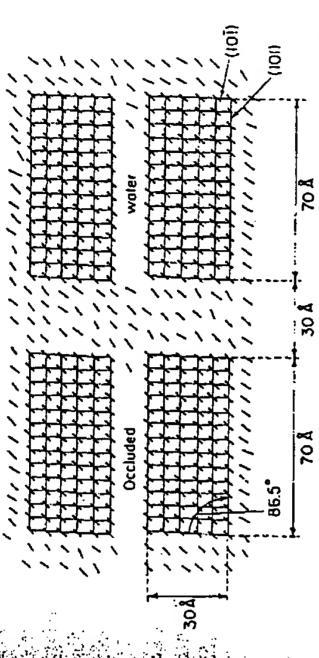


by ultrasonic entary fibrils aby) (c) Nardrop) Fig. 4.6. Electron micrographs of cellulose microfibrils and elem irradiation of the fibers in water suspension ($\times 27,000$) (R.) occurring in the primary wall of eucalypt fibers (×9,000) (from (a) purified sprucewood pulp and (b) cotton, disintegrated

ar these fibrils e values of the decrease with and of infinite ow fairly well less wide than mentary fibrils õ s noted in the s of hydrogen ften intimately further (812) .7 (235). icelles, constitute structural entities, Figure 4.6 (635). Although the hydrolysis, the elementary fibrils first tend to aggregate still improved resolution of the electron microscope, it is nestablished, that they are about 100 A wide, 30 A thick average width of the elementary fibrils have tended to those of cotton, whereas animal and algal cellulose ele may be a little wider, possibly accounting for the difference corresponding x-ray diagrams. These structural units are o and then eventually disintegrate into short fragments, m aggregated to microfibrils about 200-250 A wide by mean length. The elementary fibrils of wood cellulose may be other to an extent which permits an evaluation of how bonding between the respective surface layers, Figure

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pulps which have been alkali-treated yielding shorter micelles on hydrolysis than those which have not (414). In the former case, the micelles are about The average length of these fragments depends on the pretreatment, those the hydrolysis of cellulose suspensions is pronouncedly heterogeneous, and this assumed that the amorphous regions form the starting points of this 033, 043), rigure 4.8 100-200, which is also the average DP of the hydrolyzed pulp (70) 500-1,000 A long, corresponding to the length of molecules as the elementary norths (2/U, same width



Diagrammatic representation of a cross-section of a microfibril as an aggregation of elementary fibrils (Frey-Wyssling)



Fig. 4.8. Micelles from hydrolysed ramie cellulose, deposited on a glass surface (×47,000) (Morchead)

which removes the amorphous matter and degrades the crystalline portion attack. The degradation of the more crystalline regions first occurs in the to a more or less constant level of DP 100-200, a second phase follows, in Vicinity of the amorphous regions. After the initial phase of the hydrolysis,

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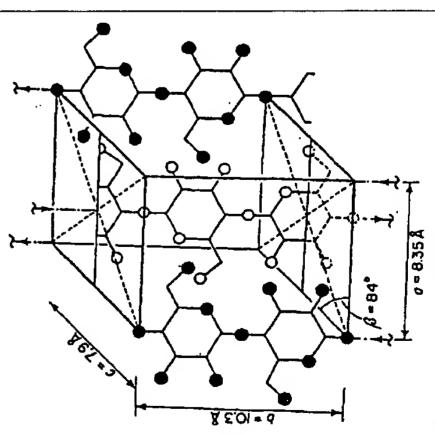
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le micellar length perfection in the icellar length and almost constant, eatment but also se of subsequent 8, 502), thus reanical influence, to introduce slip ithin the fibrils y fibrils to larger y mentioned for cotton cellulose micelles (378, cf. 719). Not only alkali tr flecting changes in the super-structure of cellulose. Mech tast for wood cellulose as for cotton. During that phase, tl and DP of the residue from the latter material remains whereas that from wood cellulose gradually decreases in m DP (559, 582). This indicates a higher degree of crystalline hydrolytic degradation and results in a lower DP level (3 planes and irregularities in the association of the elementar drying of the original cellulose induces changes in the cou such as crushing or bending of the fibers, which is known and lead to phenomena of the same type as those previous structures, cf. Chapter 3, may cause similar disturbance alkaline swelling or drying.

s been deduced tary cell with a modified (550) The detailed arrangement of the crystalline regions ha An elemen rhombic symmetry was originally proposed (629), later from x-ray data (34, 265, 325, 550, 629, 739). to a monoclinic cell with the dimensions:

$$a = 8.3 \text{ A}$$
 $b = 10.3 \text{ A}$
 $c = 7.9 \text{ A}$
 $a = 84^{\circ}$

a modification The chains are ranged parallel the a-b plane parallel to each other and are considered to hold together in as illustrated in Figure 4.9 (550). The cellulose chains are a to the b axis and have the symmetry of a digonal screw axis suggests a distortion of every second monomer (83, 590))



elementary cell cellulose (Meyer-Misch) monoclinic The Fig. 4.9.

Ö

arrangement by the length of half a monomer. Figures 4.10-12 show the planes run in opposite directions and are staggered as to their vertical by nydrogen bonding (a modification suggests hydrogen bonding between the a-b planes in the 101 direction (234)). The chains of adjacent a-b

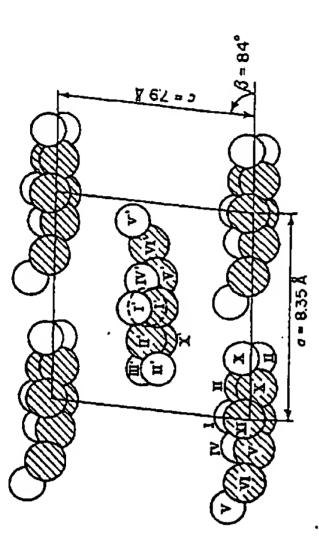
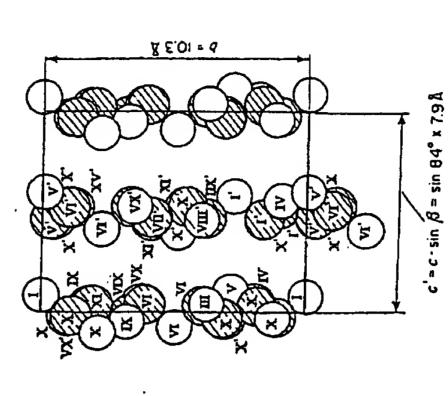


Fig. 4.10. Projection of the elementary cell of cellulose on the a-c plane perpendicular to the b-axis (Wise-Jahn)



perpendicular to the a-axis (Wise-Jahn) Fig. 4.11. Projection of the elementary ş cellulose ō

detailed arrangement of the atoms of the elementary cell in the three projections. The hydroxyl oxygen of two adjacent chains are at a distance of only 2.5 A in the direction of the a axis, allowing complete hydrogen bonding. The closest distance between the atoms belonging to different i.e. the attraction of the OH-dipoles and the permanent electric moment a-b planes is about 3.1 Å (cf. however (234)), allowing only weaker forces,

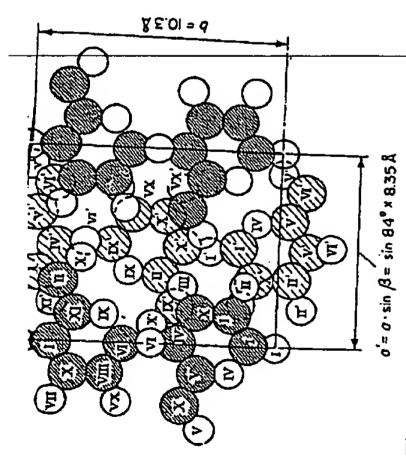


Fig. 4.12. Projection of the elementary cell of cellulose on the a-b plane perpendicular to the c-axis (Wise-Jahn)

8) indicates, that it bonds, operate er suggests, that nat an *intrachain* and every second le ring oxygen of revails also after of the 3-hydroxyl n. These recent of the classical 'lene, nylon, etc., milar folding of dvanced for disg discontinuities but itals of uniform ose molecules in r of only about e been obtained lamellar linear synthetic r-structure, mpact, observations cause only relatively small adjustments model. A more radically new concept has also been a cussion (787). Work on other polymers, such as polyethy has indicated that chain folding can occur to give crys crystals of microscopic size, similar to the single crystals of the adjacent monomer (488, 524, 797). If that bond p cellulose chain molecules in the 101 plane occurs, givin from very dilute solutions in the form of regular, co of the C-O-C groups. The strongest forces, of covaler in the direction of the b axis. Recent work (83, 590, 61 the glucose units have the C1 conformation (chair form) unit slightly twisted (20-30°). Infrared analysis furth hydrogen bond is present between the 3-hydroxyl and the in substitution reactions (143), cf. the subsequent section thickness (197, 430), and it was suggested (787) that si investigations on steric models indicate a U-turn diamete 10 Å (644), and cellulose derivatives and even cellulose har solution (e.g. 319) is an argument against such a supe for every 500 Å. The comparative rigidity of the cellul mercerization, it would probably decrease the reactivity all hydroxyls are engaged in hydrogen bonding, polymers mentioned above (644).

The crystalline structure described is valid for native cellulose. There are several polymorphous forms, of which that of regenerated cellulose is the most important. This form is also designed cellulose II in contrast to cellulose I, native cellulose. Cellulose II is formed upon regeneration of cellulose from its solid addition compounds, such as the acid, water or alkali celluloses (cf. below), as well as from solutions of addition com-

pounds (cuam or cuen solutions) or unstable substitution compounds, predominantly cellulose xanthate. Its x-ray diffraction diagram is shown in Figure 4.13 (635) in comparison to that of cellulose I. The x-ray studies have revealed (34, 114, 550) that cellulose II has a monoclinic elementary cell of the dimensions:

$$a = 8.1 \text{ A}$$

 $b = 10.3 \text{ Å}$
 $c = 9.1 \text{ Å}$

which contains four glucose monomers as in the case of cellulose I. The spatial arrangement of the chains is shown in Figure 4.14 in comparison with cellulose I. It is seen that the transformation involves a slight dis-

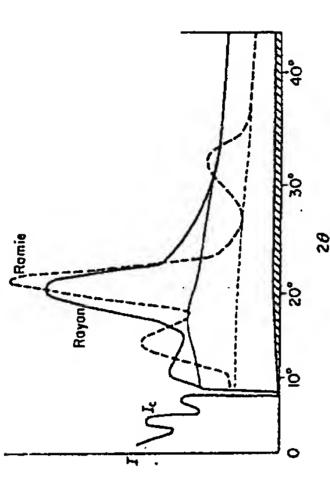


Fig. 4.13. X-ray scattering diagrams from cellulose I (ramie) and cellulose II (rayon). I_c is a reflection from a standard sample (Hermans)

the 10f direction. It is thereby noticed that the chains thus interconnected run in an opposite direction to the situation in cellulose I. Cellulose II seems to be the thermodynamically more stable form. On heating cellulose II to high temperatures in glycerol or alkali, it is converted to a new crystalline form which closely resembles cellulose I (68, 139, 326, 459, 551), but is probably a separate type, called high-temperature cellulose or cellulose IV (327, 372, 416), with an orthorhombic symmetry and the approximate dimensions:

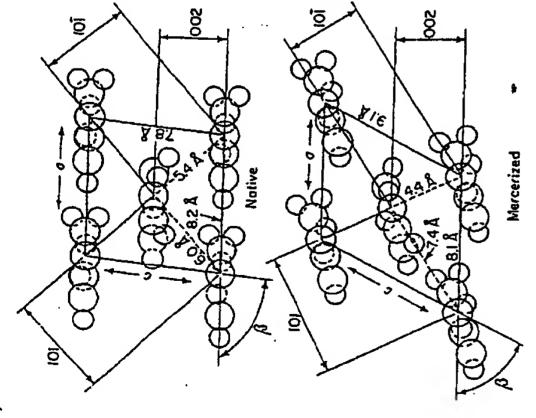
$$a = 8.1 \text{ A}$$
 $b = 10.3 \text{ A}$
 $c = 7.9 \text{ A}$
 $\theta = 90^{\circ}$

indicating a denser packing than both cellulose I and II (corresponding to a density of 1.62 as compared to 1.59 for cellulose I and II and about 1.50

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9

at temperatures le importance, as on of ammonia wwy and intination is not complete normally used in pulping processes and is therefore of litt is cellulose III, observed to form on the decomposit čellulose (326).



cellulose I and II lattices, cut at right angles to the b-axis, i.e. at right angles to the cellulose Cross-sections of the unit cells of chains 4.14. Fig.

The importance of the super-molecular structure of cellulose for its July a limited on with water, of its polarity. comparatively properties are e is influenced nd the length fraction of its hydroxyl groups is available for interaction inaccessible to chemical reagents unless the super-structure influenced not only by the average molecular length a Likewise most of its hydroxyl and acetal groups are The physical and cellulose hence remains insoluble in water in spite properties and its heterogeneous reactions is obvious. distribution, but also by the degree of crystallinity. by treatment with strong swelling agents.

REACTIVITY

ucture on the n all cellulose constitution. f reactions at various possigroups. The Like all carbohydrates, the cellulose molecule is capable of bilities of cellulose reactions are determined by its molecular its hydroxyl and acetal groups, as well as at the aldehydic end As just indicated, the influence of the super-molecular str reactivity of cellulose is profound and has to be considered reactions of heterogeneous type. However, obviously the

those deviations will be pointed out which result from the different reactions now described for cellulose will be principally possible also for the hemicelluloses, and in the section on hemicelluloses therefore only carbohydrate structure of those compounds.

agents and the acetal groups undergo hydrolysis in acid as well as in groups, oxidized to carboxyl groups or rearranged under the influence of The hydroxyl groups react with addition, substitution and oxidation sidic bonds, they will be treated together with hydrolysis under the This category of reactions alkaline medium. The aldehydic end groups can be reduced to alcohol include those of importance for the pulping processes, whereas the addition and substitution reactions are of interest mainly in connection A radical mechanism is predominant importance in the use of paper pulps. To facilitate the alkali to form either alcohol or carboxyl end groups. Since the redox and rearrangement reactions all influence the ease of hydrolysis of the gluco-A special variant of the substitution of the polyvinyl type, usually by reacting cellulose with the monomer in a involved. One addition reaction, that of carbohydrates and water, is of understanding of the various reactions, the analogous reactions with low-molecular compounds will first be treated, followed by a discussion of the reaction of cellulose in homogeneous solution, and finally the corresponding reaction in the heterogeneous two-phase system will be reactions, called grafting, involves the incorporation of copolymers, e.g. redox system, such as ceric ions (379, 710a). heading of chemical degradation reactions. with the use of dissolving pulps. described.

A. Degradation reactions

of various methyl glycopyranosides, showing the relative stability of glucosides in acid medium and of mannosides in alkaline medium. faster at low pH. The rate of reaction also varies with the type of sugar and aglycone. Table 4.18 (161, 831) gives the relative rates of hydrolysis Glycofuranosides are much more easily hydrolyzed in acid medium than Degradation of low-molecular model compounds. Hydrolysis of glycosidic bonds occurs in both acid and alkaline medium, although

Table 4.18. Relative rates of hydrolysis of various methyl glycopyranosides (161, 831)

	Acid hydrolysis	Alkaline hydrolysis
Methyl-a-glucoside	1.0	1.0
Methyl-8-glucoside	1.9	10.0
Methyl-a-mannoside	2.4	110
Methyl-8-mannoside	5.7	2.4
Methyl-a-galactoside	, ç	† ¢
Mathyl P coloctorial	7.0	5.5 6.5
Jucuisi-p-galacioside	9.3	23.0
Methyl-a-xyloside	4.5	4.8
Methyl-\(\beta\)-xyloside	9.0	23.0

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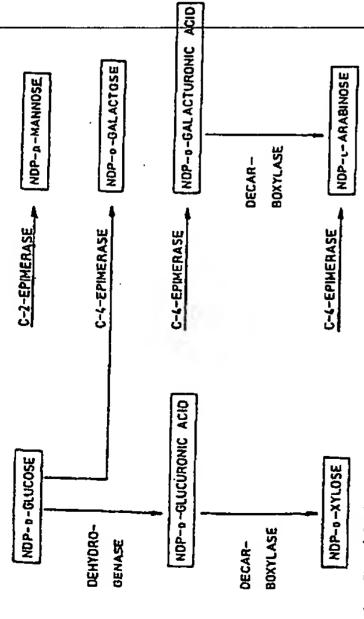
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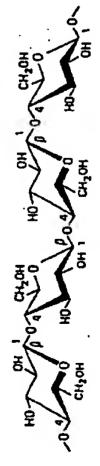


om UDP-oer UDP or Simplified representation of the formation of hemicellulose precursors fr glucose or GDP-D-glucose. Note that NDP (nucleotide diphosphate) means eit

Molecular Structure

detail, its ire is still s are the d the diopen to debate. Examples of incompletely solved problem area supermolecular state, including its crystalline and fibrillar structi exact molecular weight and polydispersity of native cellulose ar Although the chemical structure of cellulose is understood in mensions of the microfibrils.

and intermolecular hydrogen bonds. Bundles of cellulose molecules are thus Cellulose ırm intrafibrous structure and strong hydrogen bonds cellulose has a high tensile strength and is insoluble in most solvents. The physical and chemical beiose units rofibrils build up fibrils and finally cellulose fibers. As a consequence of its ordered ons. Mich clearly tics. Like 4)-linked Amylose havior of cellulose differs completely from that of starch, which demonstrates the unique influence of stereochemical characteris molecules are completely linear and have a strong tendency to for aggregated together in the form of microfibrils, in which highly (crystalline) regions alternate with less ordered (amorphous) regid Cellulose is a homopolysaccharide composed of β -D-glucopyrar D-glucopyranose units, but in starch these units are α -anomers. which are linked together by $(1 \rightarrow 4)$ -glycosidic bonds (Fig. 3-5). cellulose, the amylose component of starch consists of (1



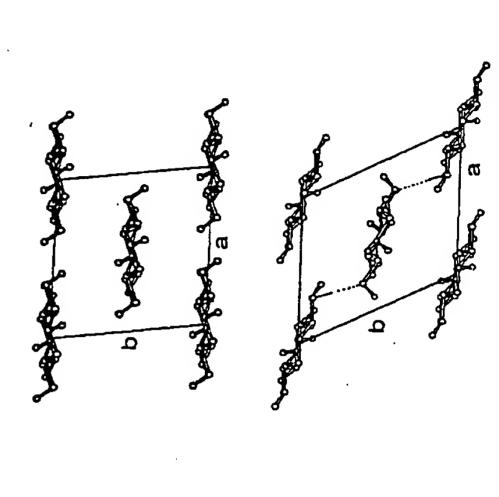
re in chair uatorially. conformation ('C₁) and the substituents HO-2, HO-3, and CH₃OH are oriented ed Structure of cellulose. Note that the β -D-glucopyranose chain units

occurs as a helix in its solid state and sometimes also in solution. Amylopec-If it, the other starch component, is also a $(1\rightarrow4)$ - α -glucan but is highly branched. The branched structure accounts for its extensive solubility, since no aggregation can take place.

3.2 Cellulose 53

four glucose residues (Figs. 3-6 and 3-7). In the chain direction (c), the The crystalline structure of cellulose has been characterized by X-ray diffraction analysis and by methods based on the absorption of polarized Infrared radiation. The unit cell of native cellulose (cellulose I) consists of repeating unit is a cellobiose residue (1.03 nm), and every glucose residue is accordingly displaced 180° with respect to its neighbors, giving cellulose a 2-fold axis. It has now been established and largely accepted that all chains they are parallel (Fig. 3-7). There are two hydrogen bonds within each in native cellulose microfibrils are oriented in the same direction, that is, cellulose chain, namely from O(6) in one glucose residue to O(2)H in the 3-8. The chains form a layer in the a-c crystallographic plane, where they are held together by hydrogen bonds from O(3) in one chain to O(6)H in the adjacent glucose and also from O(3)H to the ring oxygen, as shown in Fig. other. There are no hydrogen bonds in cellulose I between these layers, only weak van der Waal's forces in the direction of the b-axis. Native cellulose therefore has a chain lattice and a layer lattice at the same time.

3-9). The hydrogen bonds within the chains and between the chains in the Regenerated cellulose (cellulose II) (Fig. 3-6) has antiparallel chains (Fig.



Axial projections of the structures of native cellulose (cellulose I, above) and regenerated cellulose (cellulose II, below). (Reproduced from Kolpak et al., 1978, Polymer 19, 123-131, by permission of the publishers, IPC Business Press Ltd. (3.) Fig. 3-6.

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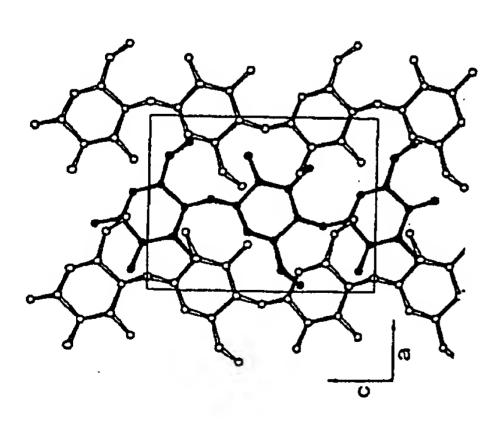


Fig. 3-7. Projection of the chains in cellulose I perpendicular to the ac plane. The center chain (black) is staggered but is parallel with the two corner chains (Gardner and Blackwell, 1974).

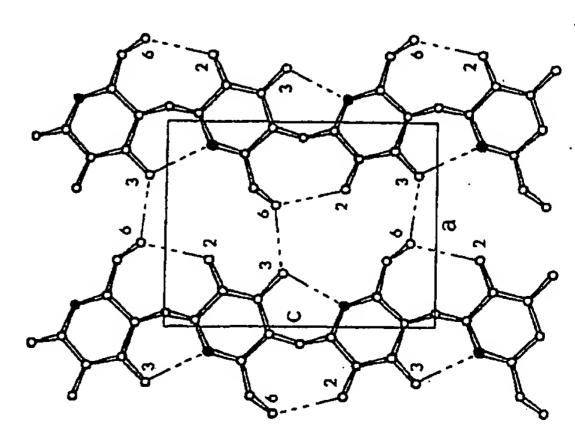


Fig. 3-8. Projection of the (O2O) plane in cellulose I, showing the hydrogen bonding network and the numbering of the atoms. Each glucose residue forms two intramolecular hydrogen bonds (03-H···05' and 06···H-02') and one intermolecular bond (06-H···03). (Slightly modified from Gardner and Blackwell, 1974.)

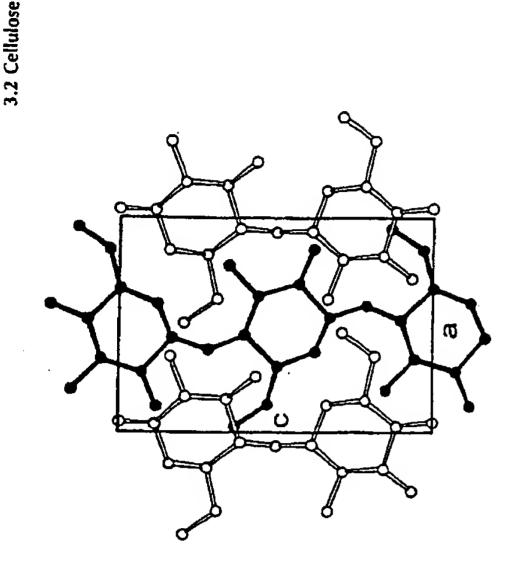


Fig. 3-9. Projection of the chains in cellulose II perpendicular to the ac plane. The center chain (black) is staggered and antiparallel to the corner chains. (Reproduced from Kolpak et al., 1978, Polymer 19, 123-131, by permission of the publishers, IPC Business Press Ltd. ©.)

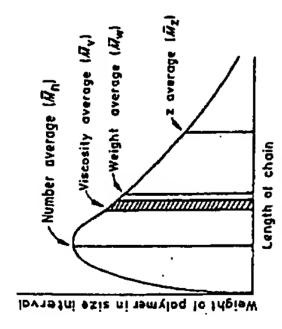
a c plane are the same as in cellulose I. In addition, there are two hydrogen bonds between a corner chain and a center chain (Fig. 3-6), namely from bonds between a corner chain and a center chain (Fig. 3-6), namely from O(2) in one chain to O(2)H in the other and also from O(3)H to 0(6). Cellulose II is formed whenever the lattice of cellulose I is destroyed, for example on swelling with strong alkali or on dissolution of cellulose. Since the strongly hydrogen bonded cellulose II is thermodynamically more stable than cellulose I, it cannot be reconverted into the latter. All naturally occerring cellulose has the structure of cellulose I. Celluloses III and IV are produced when celluloses I and II are subjected to certain chemical treatments and heating.

The proportions of ordered and disordered regions of cellulose vary considerably depending on the origin of the sample (cf. Table 9-1). Cotton cellulose is more crystalline than cellulose in wood.

32.2 The Chain Length and Polydispersity of Cellulose

The polymer properties of cellulose are usually studied in solution, using solvents, such as CED or Cadoxen (see Section 9.2). On the basis of the solution properties, conclusions can be drawn concerning the average molecular weight, polydispersity, and chain configuration. However, the

9



reights of a typical The molecular weight distribution and the average molecular w polymer (Billmeyer, 1965).

dation resulting isolation of cellulose from wood involves risk for some degra in a reduced molecular weight.

olecular weight on equilibrium ecular weight ements give an ge values differ by determining ere 162 is the so a molecular umber average lled M₂ values. asis of viscosity is a meaure of statistically as weight distribua given size measurements. For cellulose, the relationship between md and degree of polymerization (DP) is DP = M/162, wh average value of the molecular weight and some methods a data attainable by ultracentrifugation technique give so-ca Finally, $\widetilde{M}_{ extstyle v}$ refers to the molecular weight calculated on the b The distribution of molecular weights can be presented plotted against the chain length. The experimental measur weight distribution. For any polydisperse system, these avera from each other depending on the method used. The n molecular weight M, * can be measured using osmometry or Mw can be deduced from light scattering data. Sedimentat molecular weight of anhydroglucose unit. The ratio $\widetilde{M}_{w}/\overline{M}_{n}$ tion and ranges for typical polymers from 1.5-2.0 to 20-50 the number of reducing end groups. The weight average m illustrated by Fig. 3-10 where the weight of polymer of polydispersity corresponding to the width of the molecular

cellulose in its cellulose in the f about 10,000 derivatives and ons that the nae, number and rage molecular monodisperse, primary cell wall, on the other hand, which has a lower ave tive cellulose present in the secondary cell wall of plants is native state consists of about 15,000 and wood cellulose of weight average molecular weights ought to be identical. The Molecular weight measurements have shown that cotton that is; contains only molecules of one size. In such a cas polysaccharides are shown in Table 3-1. There are indicati glucose residues. Some polydispersity data on cellulose

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Polydispersity Values (Mw/Ma) of Different Polysaccharides TABLE 3-1.

Macromolecule	Source	M _w × 10-s	Mw/Ma
Cellulose nitrate	Birch	276	1 00
Cellulose nitrate	Ramie	2,4	7. 1
Amylose	Potato	A A 4	7.1
Kylan	Birch	9.0°	7.7
Xylan	Ela	0.5 0.7	22 7.4e
Amylopectin	Waxy com	, 200¢	116/
Hydrolyzed amylopectin	Waxy corn	158	756
Glycogen	Sweet com	190	15
Hydrolyzed glycogen	Sweet corn	20 _b	/t 9
Glycogen	Rabbit liver	390	6.69

From Goring (1962)

LAW

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by light scattering.

^e \vec{M}_n by viscometry from $[\eta] = 0.0091$ DP.

d Calculated from fractionation data.

^e M_n by osmometry.

'Mn from the alkali number.

 \widetilde{M}_{ww} from sedimentation and diffusion used instead of \widetilde{M}_{n} ; usually $\widetilde{M}_{w} > \widetilde{M}_{ww} > \widetilde{M}_{n}$.

weight, is evidently polydisperse, being similar in this respect to the hemicelluloses.

The Configuration of Cellulose Molecules 3.2.3

Based on properties in solution such as intrinsic viscosity and sedimentation and diffusion rates, conclusions can be drawn concerning the polymer configuration. Like most of the synthetic polymers, such as polystyrene, cellulose in solution belongs to a group of linear, randomly celling polymers. This means that the molecules have no preferred structure in solution in contrast to amylose and some protein mofecules which can adopt helical conformations. Cellulose differs distinctly from synthetic polymers and from lignin in some of its polymer properties. Typical of its solutions are the comparatively high viscosities and low sedimentation and diffusion coefficients (Tables 3-2 and 3-3).

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2/ Any linear polymer molecule, even a reasonably stiff rod, will coil randomly, provided the chain is sufficiently long. In addition to the size of the monomer units, the tendency for coiling is affected by the forces between the units as well as the interaction between the polymer and the solvent. tance (R). For a polydisperse polymer the root-mean-square average of R by the interaction of the solvent. The better the solvent the more the polymer One measure of the stiffness of a polymer molecule is the end-to-end dis- $(\overline{R}^2)^{1/2}$ is used. R is affected by the properties of the polymer itself as well as

ially adopted in the relative molecular *The SI system (Système International d'Unités) recommends the term mass instead of molecular weight, but because the SI term is not yet univer polymer chemistry the latter term is used throughout this book.

RELATED PROCEEDINGS APPENDIX

(NONE)

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